# Europäisches Patentamt European Patent Office Office européen des brevets



(11) **EP 1 270 656 A1** 

(12)

# **EUROPEAN PATENT APPLICATION**

(43) Date of publication: 02.01.2003 Bulletin 2003/01

(51) Int CI.7: **C08K 3/04**, C08L 21/00, B60C 1/00

(21) Application number: 02014166.9

(22) Date of filing: 25.06.2002

(84) Designated Contracting States:

AT BE CH CY DE DK ES FI FR GB GR IE IT LI LU MC NL PT SE TR
Designated Extension States:

AL LT LV MK RO SI

AL EI EV MIKTIO OI

(30) Priority: 29.06.2001 JP 2001199025

(71) Applicant: SUMITOMO RUBBER INDUSTRIES LIMITED

Kobe-shi, Hyogo-ken (JP)

(72) Inventor: Mizuno, Yoichi, c/oSumitomo Rubber Industries Ltd. Kobe-shi, Hyogo-ken (JP)

(74) Representative:

Manitz, Finsterwald & Partner GbR Postfach 31 02 20 80102 München (DE)

## (54) Rubber composition for tread base and pneumatic tire

(57) The present invention provides a rubber composition for base tread having reduced heat build-up characteristics and excellent reinforcing property, and a pneumatic tire using the rubber composition for the base tread. The rubber composition for base tread of the present invention comprises 30 to 40 parts by weight of carbon black having iodine adsorption amount of at least 115 m<sup>2</sup>/g, 5 to 10 parts by weight of silica and 1.2 to 2.2

parts by weight of sulfur based on 100 parts by weight of a rubber component, wherein the total amount of the carbon black and the silica is at most 45 parts by weight. The pneumatic tire of the present invention is obtained by using 20 to 80 % by volume of a base tread comprising the rubber composition of Claim 1 based on the total volume of a tread.

#### Description

5

10

20

25

30

40

45

55

**[0001]** The present invention relates to a rubber composition for base tread and a pneumatic tire using the same. Particularly, the present invention relates to a rubber composition for base tread having reduced heat build-up characteristics and excellent reinforcing property, and a pneumatic tire using the rubber composition for base tread.

**[0002]** In recent years, demand for improvement of tire life is increasing, and there are developments such as improvement of abrasion resistance of cap tread rubber and increase of tread width. However, such attempts are now resulting in problems that a tire has increased heat build-up characteristics to cause separation of tread from belt owing to the heat build-up, and that rolling resistance of a tire is increased.

**[0003]** As a means for alleviating these problems, a general technique is to form a two-layer structure tread comprising cap and base, using a low heat build-up rubber for the base tread. However, reinforcing property of a usual low heat build-up rubber is poor, and therefore abrasion resistance decreases when the base tread is exposed at a late stage of abrasion, causing poor abrasion appearance (chipping) or crack at the bottom of the groove. Therefore, the low heat build-up rubber can be only used in such an amount that it is not exposed to the surface even if abrasion occurs, and lowering effect on heat build-up remains small.

[0004] In order to meet the demand for long life tires, primal task is to develop a rubber composition having reduced heat build-up characteristics and high reinforcing property, achieving a practical use of low heat build-up tire having a structure with increased base volume.

[0005] In order to achieve low heat build-up characteristics, carbon or silica having large particle diameters has been added to conventional base tread compounds. However, when the volume of base tread is increased to such an extent that the base tread is exposed at the last stage of abrasion for the purpose of achieving further reduced heat build-up characteristics and increased fuel efficiency, durability has been decreased, and such increase was difficult. Therefore, these conventional tread compounds were not easily applied to bus or truck tires used under particularly severe conditions.

[0006] As a result of studies to solve the above problems, a compound having both reduced heat build-up characteristics and high reinforcing ability has been found by using carbon black having a large specific surface area, which provides excellent abrasion resistance and elongation at break, together with silica which provides reduced heat build-up characteristics and excellent elongation at break, and by optimizing these amounts and the amount of sulfur.

[0007] Furthermore, the cap/base ratio of the tread has also been optimized.

[0008] That is, the present invention relates to a rubber composition for base tread comprising, 30 to 40 parts by weight of carbon black having iodine adsorption amount of at least 115 m<sup>2</sup>/g, 5 to 10 parts by weight of silica and 1.2 to 2.2 parts by weight of sulfur based on 100 parts by weight of a rubber component, wherein the total amount of the carbon black and the silica is at most 45 parts by weight.

[0009] The present invention also relates to a pneumatic tire obtained by using 20 to 80 % by volume of a base tread comprising the rubber composition of Claim 1 based on the total volume of a tread.

**[0010]** The rubber composition of the present invention is obtained by compounding particular amounts of carbon black having a large specific surface area, silica and sulfur to a rubber component.

[0011] As the rubber component, natural rubber (NR) and/or isoprene rubber (IR) are preferable from the viewpoint of heat build-up characteristics. It is more preferable that NR and/or IR are used in a total amount of at least 90 % by weight.

**[0012]** Examples of other useful rubber components include butadiene rubber (BR), styrene-butadiene rubber (SBR), isoprene rubber (IR), butyl rubber (IIR), acrylonitrile-butadiene rubber, chloroprene rubber, ethylene-propylene rubber, styrene-isoprene rubber, styrene-isoprene-butadiene rubber, isoprene-butadiene rubber, chlorosulfonated polyethylene, acrylic rubber, epichlorohydrine rubber, polysulfide rubber, silicone rubber, fluorine rubber, urethane rubber, bromides of isobutylene-p-methyl styrene copolymer and the like.

**[0013]** Referring to carbon black, one having an iodine adsorption amount of at least 115 m<sup>2</sup>/g is compounded. It is preferable that the lower limit of iodine adsorption amount is 120 m<sup>2</sup>/g and the upper limit of iodine adsorption amount is 180 m<sup>2</sup>/g. When the iodine adsorption amount is less than 115 m<sup>2</sup>/g, abrasion resistance, abrasion appearance and elongation at break are inferior. On the other hand, when the iodine adsorption amount is more than 180 m<sup>2</sup>/g, heat build-up characteristics tend to increase.

[0014] Concrete examples of carbon black are N110, N220, N219, N134 and the like.

[0015] The amount of carbon black used in the present invention is 30 to 40 parts by weight based on 100 parts of the rubber component. When the amount of carbon black is less than 30 parts by weight, abrasion resistance is insufficient. When the amount is more than 40 parts by weight, heat build-up characteristics increase. Preferably, the upper limit of the amount of carbon black is 38 parts by weight and the lower limit of the amount of carbon black is 32 parts by weight.

**[0016]** With respect to silica, one having a BET adsorption amount of 100 to 240 m<sup>2</sup>/g is preferable. When the BET adsorption amount of silica is less than 100 m<sup>2</sup>/g, abrasion resistance and reinforcing properties such as chipping

resistance tend to be inferior. When the BET adsorption amount of silica is more than 240 m<sup>2</sup>/g, heat build-up characteristics tend to increase.

[0017] The amount of silica is 5 to 10 parts by weight based on 100 parts by weight of the rubber component. When the amount of silica is less than 5 parts by weight, low heat build-up characteristics and chipping resistance (elongation at break) are inferior. When the amount is more than 10 parts by weight, abrasion resistance decreases. Preferably, the upper limit of the amount of silica is 8 parts by weight and the lower limit of the amount of silica is 5 parts by weight. [0018] The total amount of carbon black and silica is at most 45 parts by weight, preferably at most 43 parts by weight based on 100 parts by weight of the rubber component. When the total amount is more than 45 parts by weight, heat build-up characteristics increase. The total amount is preferably at least 35 parts by weight, more preferably at least 38 parts by weight based on 100 parts by weight of the rubber component. When the total amount is less than 35 parts by weight, abrasion resistance tends to decrease.

[0019] The amount of sulfur is 1.2 to 2.2 parts by weight based on 100 parts by weight of the rubber component. When the amount of sulfur is less than 1.2 parts by weight, heat build-up characteristics increase. When the amount of sulfur is more than 2.2 parts by weight, abrasion resistance and chipping resistance decrease. Preferably, the upper limit of the amount of sulfur is 2.0 parts by weight and the lower limit of the amount of the sulfur is 1.4 parts by weight. [0020] The rubber composition for base tread of the present invention may contain a usual amount of additives or components commonly used for producing rubber compositions for tire tread, if necessary, in addition to the rubber component, carbon black, silica and sulfur. Concrete examples of the additives and the components are process oils such as paraffin process oil, naphthene process oil and aromatic process oil; vulcanization accelerators such as guanidine, aldehyde-amine, thiourea, thiuram, dithiocarbamate and xandate compounds; cross-linking agents such as radical initiators including organic peroxide compounds and azo compouds, oxime compounds, nitroso compounds and polyamine compounds; reinforcing agents such as high impact polystyrene resins and phenol-formaldehyde resins; antioxidants such as amine derivatives including diphenylamine derivatives or p-phenylenediamine derivatives, quinoline derivatives, hydroquinone derivatives, monophenols, diphenols, thiobisphenols, hinderedphenols and phosphorous acid ester; waxes; stearic acids; zinc oxides; softeners; and plasticizers.

**[0021]** The pneumatic tire of the present invention has a tread which comprises a base tread (inner layer) and cap tread (surface layer). A tread having such a structure can be prepared by a method which comprises kneading the respective components of base tread and cap tread by using usual processing apparatus such as a roll, a Banbury mixer or a kneader to form each sheet, and laminating the sheets to give a pre-determined shape, or by a method which comprises introducing the components into at least two extruders respectively, so that two layers are formed at the end of head of the extruders. The pneumatic tire of the present invention has a base tread comprising the above rubber composition for base tread.

**[0022]** The volume ratio of cap tread to base tread (cap tread/base tread) is 80/20 to 20/80. When the volume of the base tread is less than 20 % by volume, improvement of heat build-up characteristics and rolling resistance is insufficient. When the volume of the base tread is more than 80 % by volume, the tire to be obtained has remarkably decreased abrasion resistance, and costs are also increased. Preferably, the upper limit of the volume of the base tread is 70 % by volume and the lower limit is 30 % by volume.

**[0023]** The loss tangent ( $\tan \delta$ ) of the entire tread is preferably 0.080 to 0.120. The  $\tan \delta$  of the entire tread is calculated as follows:

 $\tan \delta$  of the entire tread = (tan  $\delta$  of cap tread)  $\times$  (volume ratio of cap tread)

+ (tan  $\delta$  of base tread)  $\times$  (volume ratio of base tread)

[0024] When the  $\tan \delta$  of the entire tread is less than 0.080, reinforcing ability tends to be inferior. When the  $\tan \delta$  of the entire tread is more than 0.120, improvement of heat build-up characteristics tends to be insufficient. More preferably, the upper limit thereof is 0.100 and the lower limit thereof is 0.085.

## **EXAMPLES**

[0025] The present invention is explained in detail based on Examples below, but not limited thereto. "Part(s)" and "%" in the following examples mean "part(s) by weight" and "% by weight", respectively, unless otherwise specified.

55

50

10

20

25

30

40

## EXAMPLES 1 to 8 and COMPARATIVE EXAMPLES 1 to 12 (RUBBER COMPOSITIONS)

(Compounded components)

## 5 [0026]

15

20

30

35

40

45

NR: RSS#3

BR: BR150B available from Ube Industries, Ltd.

Carbon black 1: Seast 9 (N110) available from Tokai Carbon Co., Ltd. (iodine adsorption amount: 142 m<sup>2</sup>/g)

Carbon black 2: DIABLACK I (N220) available from Mitsubishi Chemical Corporation (iodine adsorption amount: 118 m²/g)

Carbon black 3: Seast N (N330) available from Tokai Carbon Co., Ltd. (iodine adsorption amount: 70 m<sup>2</sup>/g)

Silica: Ultrasil VN 3 available from Degussa Co.

Silane coupling agent: bis(3-triethoxysilylpropyl)tetrasulfene (Si69) available from Degussa Co.

Wax: Sannowax available from Ohuchi Shinko Kagaku Kogyo Co. Ltd.

Antioxidant: Ozonone 6C available from Seiko Chemicals Co., Ltd.

Stearic acid: KIRI available from NOF Corporation

Zinc oxide: Ginrei R available from Toho Zinc Co. Ltd.

Sulfur: sulfur available from Tsurumi Chemicals Co., Ltd.

Vulcanization accelerator: Nocceler NS available from Ohuchi Shinko Kagaku Kogyo Co. Ltd. (N-tert-butyl-2- ben-zothiazolylsulfenamide)

(Preparation method)

25 [0027] The basic compound shown in Table 1 was compounded with each compound shown in Tables 2 and 3 except for sulfur and vulcanization accelerator. The mixture was kneaded by using a Banbury mixer at about 150°C for five minutes. To the obtained rubber composition were added sulfur and vulcanization accelerator, and the composition was kneaded at about 80°C for five minutes by a twin-screw open roll

[0028] The obtained rubber composition for base tread was molded and vulcanized at 150°C for 30 minutes under a pressure of 20 kgf to prepare a truck tire having a size of 11R22.5. For the cap tread, an all-season tread rubber was used (compound: 80 parts of the NR (RSS#3), 20 parts of the BR (BR150B), 54 parts of the carbon black (N110), 2 parts of the antioxidant, 1 part of the wax, 2 parts of stearic acid, 5 parts of zinc oxide, 1.2 parts of sulfur and 1.5 parts of the vulcanization accelerator). Test pieces were prepared from the base tread rubber composition of the new tire, and evaluated as follows:

(Test method)

1) Loss tangent (elasticity test)

[0029] Test pieces were prepared from the base tread rubber composition of the new tire. Loss tangent (tan  $\delta$ ) of each test piece at 60°C was measured by using a viscoelastometer made by Iwamoto Corporation under the condition of a frequency of 10 Hz and a dynamic strain of 1.0 %. The smaller the tan  $\delta$ , the lower and more excellent the heat build-up characteristics, and the more excellent the rolling resistance. When the tan  $\delta$  is at most 0.100, it is evaluated that heat build-up characteristics is sufficiently low.

2) Abrasion resistance test

[0030] Test pieces were prepared from the base tread rubber composition of the new tire. Using a Lambourne abrasion tester made by Iwamoto Corporation, volume loss (abrasion amount) of each test piece was measured under the condition of a surface rotation speed of 50 m/minute, a load of 5.0 kg, a drop rate of sand of 15 g/minute, a slip ratio of 50 % and a testing time of 4 minutes. The measured volume loss was represented as an index to the volume loss value of Comparative Example 1 as 100 according to the following equation (abrasion resistance index). The larger the index, the more excellent the abrasion resistance. When the index is at least 110, it is evaluated that abrasion resistance is excellent.

55

(Lambourne abrasion index) = (volume loss value of Comparative

Example 1) ÷ (volume loss value of each compound) × 100

3) Test of elongation at break (tensile test)

**[0031]** Tensile test was carried out by using a No. 3 dumbbell specimen prepared from the base tread rubber composition of the new tire according to JIS-K6251 to measure elongation at break  $E_B$  (%).  $E_B$  was represented as an index to the value of elongation at break of Comparative Example 1 as 100 according to the following equation (index of elongation at break). The larger the index, the more excellent the elongation at break and chipping resistance in particular. When the index is at least 110, it is evaluated that chipping resistance is excellent.

(index of elongation at break) = (elongation at break of each compound) ÷

(elongation at break of Comparative Example 1)  $\times$  100

(Test results)

5

10

15

20

25

30

35

40

[0032] Test results are shown in Tables 2 and 3. In Comparative Examples 1 to 4 where carbon black or silica was solely compounded as a filler, desired balanced performances could not be obtained.

[0033] In Comparative Example 5 where carbon black having a low iodine adsorption amount was used, desired balanced performances could not be obtained.

[0034] Contrary to these, in Examples 1 and 2 where carbon black having a low iodine adsorption amount and silica were compounded as fillers, desired balanced performances could be obtained.

[0035] Comparison between Examples 2 to 4 and Comparative Examples 6 and 7 shows that it is preferable to compound silica in an amount of 5 to 10 parts.

**[0036]** Comparison between Examples 2, 5 and 6 and Comparative Examples 8 to 10 shows that it is preferable to compound carbon black in an amount of 30 to 40 parts and that the total amount of silica and carbon black is at most 45 parts by weight.

[0037] Comparison between Examples 2, 7 and 8 and Comparative Examples 11 and 12 shows that it is preferable to compound sulfur in an amount of 1.2 to 2.2 parts.

TABLE 1

IADI	<u>-</u>
Compounded component	Amount (Part by weight)
NR	100
Antioxidant	2
Wax	2
Stearic acid	2
Zinc oxide	5
Vulcanization accelerator	1.5

45

50

TABLE 2

		Ex. 1	Ex. 1 Ex. 2	Com. Ex. 1	Com. Ex. 2	Com. Ex. 3	Com. Ex. 4	Com. Ex. 5	Ex. 3	Ex. 4	Com. Com.	Com.
	Carbon black 1	I	35	1	,	43	ı	1	35	35	35	35
	Carbon black 2	35	ı	1	43	i	I	ı	I	1	ı	ı
Compound	Compound Carbon black 3	I	i	43	i	ı	ı	35	I	ı	I	ı
(part by	Silica	∞	∞	ı	I	1	<del>2</del> 8 4	∞	rV	10	က	12
weignt)	Coupling agent	0.8	8.0	I	ι	1	4.3	8.0	0.5	-	0.3	1.2
	Sulfur	1.6	1.6	1.6	1.6	1.6	1.6	1.6	1.6	1.6	1.6	1.6
	Carbon black + Silica	43	43	43	43	43	43	43	40	45	38	47
	1) Loss tangent	0.075	0.075 0.087	0.074	0.074 0.090 0.106 0.060 0.065	0.106	0.060	0.065	0.085	0.094	0.080 0.102	0.102
Properties of rubber	2) Abrasion resistance index	110	117	100	112	119	86	26	114	121	109	124
	3) Index of elongation at break	114	117	100	109	114	132	107	121	114	124	112

5	
10	
15	
20	
25	
30	
35	
40	
45	
50	

55

TABLE 3

		Ex. 5 Ex. 6	Ex. 6	Com. Com. Com. Ex. 8 Ex. 9 Ex. 10	Com. Ex. 9	Com. Ex. 10	Ex. 7 Ex. 8	Ex. 8	Com. Com. Ex. 11Ex. 12	Com. Ex. 12
	Carbon black 1	30	40	25	40	45	35	35	35	35
	Carbon black 2	ŀ	I	1	ı	ı	ı	1	1	ł
	Carbon black 3	ı	1	1	1	ı	ı	ı	ı	ı
Compound	Silica	∞	ស	œ	∞	∞	∞	<b>∞</b>	œ	œ
(part for more true)	Coupling agent	0.8	0.5	0.8	0.8	0.8	0.8	0.8	0.8	8.0
	Sulfur	1.6	1.6	1.6	1.6	1.6	1.2	2.2	_	2.5
	Carbon black + Silica	38	45	33	48	53	43	43	43	43
	1) Loss tangent	0.072	0.072 0.097	0.060 0.107 0.121	0.107	0.121	0.097	0.097 0.080	0.104	0.072
Properties of mibber	2) Abrasion resistance index	114	122	106	126	131	124	110	130	104
	3) Index of elongation at break	121	117	129	113	107	124	112	128	106

## EXAMPLES 9 to 14 and COMPARATIVE EXAMPLES 13 to 15 (CHARACTERISTICS OF TIRE)

(Preparation method)

- <sup>5</sup> [0038] Truck tires having a size of 11R22.5 were prepared, whose tread comprises cap tread and base tread in each volume ratio shown in Table 4. Vulcanization was carried out under the condition of 150°C and a pressure of 20 kgf for 30 minutes. An all-season tread rubber was used for the cap tread (index of elongation at break: 130, loss tangent: 0.130 in the above evaluation).
- 10 (Evaluation method)

15

20

25

30

35

40

45

- 4) Rolling resistance
- [0039] Rolling resistance of each tire was measured under the condition of a speed of 80 km/h, an air pressure of 800 kPa and a load of 30 kN. Each value was represented as an index to the rolling resistance value of Comparative Example 1 as 100 according to the following equation (rolling resistance index). The larger the index, the more excellent the rolling resistance and fuel efficiency. When the index is at least 103, it is evaluated that fuel efficiency is excellent.

(rolling resistance index) = (rolling resistance value of Comparative

Example 1) ÷ (rolling resistance value of each compound) × 100

5) Tire heat build-up test

**[0040]** Each tire was mounted on a 10 ton truck and run for 10 minutes at a speed of about 80 km/h to measure temperature of the upper part of the breaker edge. Each heat build-up temperature value was represented as an index to the heat build-up temperature value of Comparative Example 1 as 100 according to the following equation (heat build-up index). The larger the index, the lower the heat build-up characteristics and the more preferable. When the index is at least 103, it is evaluated that heat build-up characteristics is sufficiently low.

(heat build-up index) = (heat build-up temperature value of Comparative

Example 1)  $\div$  (heat build-up temperature value of each compound)  $\times$  100

6) Abrasion appearance of base tread

**[0041]** Each tire was mounted on a 10 ton truck and run for 200,000 km, so that the base tread was exposed. Abrasion appearances (chipping levels) were compared. The number of flaws with a length of at least 1 mm was counted per 100 cm<sup>2</sup> of the base tread surface. When the number of flaws is at most 10, in particular at most 5, it is evaluated that the appearance level is equal to cap tread and there is no problem.

(Evaluation results)

[0042] Results are shown in Table 4.

**[0043]** Table 4 shows that heat build-up characteristics are large in Comparative Example 13 without base tread, Comparative Example 15 where the base tread had a large loss tangent because too large an amount of carbon black having a large iodine adsorption amount was compounded, and in Example 14 whose base tread ratio was small, because the loss tangent of the entire tread is too large.

[0044] In addition, it is found that there is a problem with abrasion appearance of base tread in Comparative Example 14 where the base tread had a small loss tangent because carbon black having a small iodine adsorption amount was compounded.

TABLE 4

	Ex. 9	Ex. 9 Ex. 10 Ex. 11 Ex. 12 Ex. 13 Ex. 14	Ex. 11	Ex. 12	Ex. 13	Ex. 14	Com. Ex. 13	Com. Com. Com. Ex. 13 Ex. 14 Ex. 15	Com. Ex. 15
Cap tread	9	09	80	40	20	90	100	09	09
Rubber composition of Com. Ex. 1	I	ı	ı	ı	I	1	1	40	I
Rubber composition of Com. Ex. 3	ł	ı	1	1	i	1	I	ı	40
Dase u cau Rubber composition of EX. 1	40	1	20	9	80	10	I	ı	ı
Rubber composition of EX. 2	ı	40	I	ı	1	i	I	i	ı
tanð:(tanð of cap) × (volume ratio of cap) + (tanð of base) × (volume ratio of base)	0.108	0.113	0.119	0.097	0.086	0.108 0.113 0.119 0.097 0.086 0.125 0.130 0.108 0.120	0.130	0.108	0.120
4) Rolling resistance index	107	104	105	108	110	101	100	106	102
Properties 5) Heat build-up index of tire	106	105	104	107	111	101	100	104	102
6) Abrasion appealance of base tread (number of flaws)	5	77	7	9	7	7	2	21	4

**[0045]** The rubber composition for base tread of the present invention has reduced heat build-up characteristics and excellent reinforcing property. Therefore, abrasion resistance is not lowered even if base tread is exposed at the last stage of abrasion. Accordingly, it is possible to increase the volume of the base tread compared with conventional cases, and in addition, heat build-up characteristics of tire can be reduced.

#### Claims

5

15

20

25

30

35

40

45

50

55

1. A rubber composition for base tread comprising, 30 to 40 parts by weight of carbon black having iodine adsorption amount of at least 115 m<sup>2</sup>/g,

5 to 10 parts by weight of silica and

1.2 to 2.2 parts by weight of sulfur

based on 100 parts by weight of a rubber component,

wherein the total amount of the carbon black and the silica is at most 45 parts by weight.

2. A pneumatic tire obtained by using 20 to 80 % by volume of a base tread comprising the rubber composition of Claim 1 based on the total volume of a tread.



# **EUROPEAN SEARCH REPORT**

**Application Number** EP 02 01 4166

	DOCUMENTS CONSIDERED  Citation of decument with indication		Relevant	CLASSIFICATION OF THE
Category	of relievant passages	, who appropriate,	to claim	APPLICATION (Int.CI.7)
Х	US 5 225 011 A (ODA KEIJ 6 July 1993 (1993-07-06) * abstract; claims; figu	,	1,2	C08K3/04 C08L21/00 B60C1/00
X	EP 0 738 614 A (GOODYEAR 23 October 1996 (1996-10 * abstract; claims; tabl * page 7, line 33 - line * page 5, line 4 - line	-23) e 1 * 52 *	1,2	
x	EP 0 708 136 A (UNIROYAL 24 April 1996 (1996-04-2 * abstract; claims; exam * page 5, line 12 - line	4) ple 4 *	1,2	
x	EP 0 942 041 A (SUMITOMO 15 September 1999 (1999- * claims; tables 2,3 * * page 3, line 44 *		1	
				TECHNICAL FIELDS SEARCHED (Int.Cl.7)
				B60C   C08K
1	The present search report has been drag	wn up for all claims		
	Place of search	Date of completion of the search		Examiner
	THE HAGUE	26 September 20	02 Met	tler, R-M
X : partic Y : partic docu A : techr	ATEGORY OF CITED DOCUMENTS  cularly relevant if taken alone cularly relevant if combined with another ment of the same category hological background	E ; earlier patent of after the filing of D : document cited L : document cited	iple underlying the document, but publicate d in the application d for other reasons	nvention shed on, or
O: non-	nological background -written disclosure mediate document		same patent tamily	

## ANNEX TO THE EUROPEAN SEARCH REPORT ON EUROPEAN PATENT APPLICATION NO.

EP 02 01 4166

This annex lists the patent family members relating to the patent documents cited in the above-mentioned European search report. The members are as contained in the European Patent Office EDP file on The European Patent Office is in no way liable for these particulars which are merely given for the purpose of information.

26-09-2002

	Patent docume cited in search re		Publication date		Patent fan member(		Publication date
US	5225011	А	06-07-1993	JP JP	61287802 61287945		18-12-1986 18-12-1986
EP	0738614	А	23-10-1996	AU AU BR CA CN EP JP TR	694334 5078996 9601411 2171393 1140668 0738614 8302072 970158	A A A1 A,B A1 A	16-07-1998 31-10-1996 13-01-1998 22-10-1996 22-01-1997 23-10-1996 19-11-1996 21-03-1997
EP	0708136	A	24-04-1996	DE AT DE EP		T D1	25-04-1996 15-03-1999 08-04-1999 24-04-1996
ΕP	0942041	A	15-09-1999	JP EP US	11255966 0942041 6378584	A A1 B1	21-09-1999 15-09-1999 30-04-2002

OFIM P0459

For more details about this annex : see Official Journal of the European Patent Office, No. 12/82